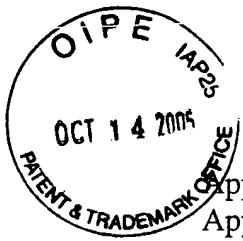


IN THE UNITED STATES PATENT AND TRADEMARK OFFICE



Appl. No. : 10/678,459  
Applicant : Kenneth Paul Zarnoch et al.  
Filed : October 3, 2003  
TC/A.U. : 1712  
Examiner : David J. Buttner  
  
Assignee Docket No. : 132177-1  
Attorney Docket No. : G21-0003  
Customer No. : 23413

Via Facsimile (571) 273-8300, TC Group 1700  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

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Date: Oct. 4, 2005

Kenneth Paul Zarnoch  
Kenneth Paul Zarnoch

Date: Oct 4, 2005

John Robert Campbell

Date: Oct. 4, 2005

Glen David Merfeld  
Glen David Merfeld

Date: \_\_\_\_\_

John Rude

Date: \_\_\_\_\_

Prameela Susarla

Date: Oct 4, 2005

Michael Alan Vallance  
Michael Alan Vallance

Date: \_\_\_\_\_

Gary William Yeager

132177-1  
(G21-0003)



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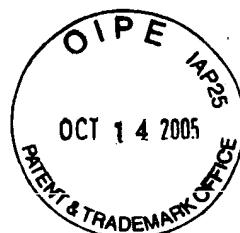
Date: \_\_\_\_\_ John Rude

Date: OCT 5, 2005 Prameela Susarla

Date: \_\_\_\_\_ Michael Alan Vallance

Date: \_\_\_\_\_ Gary William Yeager

132177-1  
(G21-0003)



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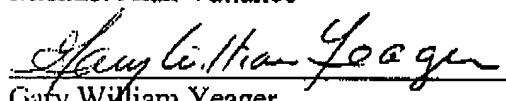
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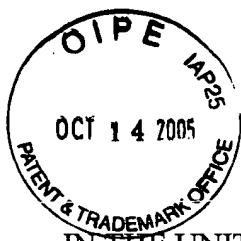
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132177-1  
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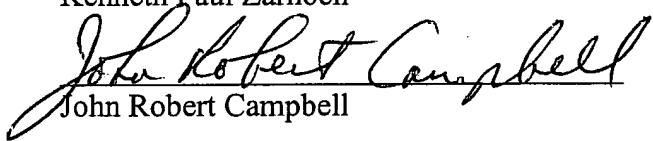
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4. In Exhibit 2, particular attention is called to "Formulation 12." The composition of Formulation 12 is given in the last column of the formulation table on Exhibit 2, page 2. In Formulation 12, "PPO-MA(0.12)" is a methacrylate-capped poly(2,6-dimethyl-1,4-phenylene ether) having an intrinsic viscosity of about 0.12 deciliters per gram measured at 25°C in chloroform; and "TMPTMA" is trimethylolpropane trimethacrylate. The component names "Diallylphthalate", "Dicumyl Peroxide", "t-butyl catechol", "Lecithin", "Esterified Montanic Acid", "Montanic acid", "Dibromostyrene", "Antimony pentoxide", "Carbon black", "Silane coupling agent", and "Fused silica" are assumed to be self-explanatory. Components for which names and amounts are in faint text (i.e., Dibromostyrene, Antimony pentoxide, Carbon black, Silane coupling agent, and Fused Silica) are constant for all formulations.

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and R<sup>4</sup> is methyl, and m is about 30; K has the structure (-Y-C(R<sup>6</sup>)=C(R<sup>7</sup>)(R<sup>8</sup>)), where Y is (-C(=O)-), R<sup>6</sup> is methyl, and R<sup>7</sup> and R<sup>8</sup> are hydrogen. The claim 1 limitation, "an olefinically unsaturated monomer" is satisfied, collectively, by "Diallylphthalate", "TMPTMA", and "Dibromostyrene". The claim 1 limitation, "about 0.2 to about 5 part by weight of a curing initiator per 100 parts by weight total of the functionalized poly(arylene ether) and the olefinically unsaturated monomer" is satisfied by "Dicumyl Peroxide" at 0.052 parts by weight, which corresponds to 2.36 parts by weight per 100 parts by weight total of "PPO-MA(0.12)", "Diallylphthalate", "TMPTMA", and "Dibromostyrene". The claim 1 limitation "about 0.005 to about 1 part by weight of a curing inhibitor per 100 parts by weight total of the functionalized poly(arylene ether) and the olefinically unsaturated monomer" is satisfied by "t-butyl catechol" at 0.05 parts by weight, which corresponds to 0.227 parts by weight per 100 parts by weight total of "PPO-MA(0.12)", "Diallylphthalate", "TMPTMA", and "Dibromostyrene". The limitation, "wherein the weight ratio of the curing initiator to the curing inhibitor is about 1.2:1 to about 50:1" is satisfied by the weight ratio of "Dicumyl Peroxide" to "t-butyl catechol", which is 0.52:0.05, or 10.4:1.

6. The Exhibit 1 spreadsheet was originally attached to the Exhibit 3 patent disclosure letter. See page 5 of Exhibit 3, where "(021127,Susarla) Spiral flow study for Disclosure.xls" is listed as an "ATTACHED FILE". The redacted "DOCKET DATE" on the first page of Exhibit 3, the redacted "Date Invention Conceived" on the second page of Exhibit 3, and the redacted date associated with the question, "Has your invention been reduced to practice?" on page 4 of Exhibit 3, are all prior to July 25, 2003. In other words, "Formulation 12" was designed and prepared prior to July 25, 2003. Therefore, conception and actual reduction to practice of the present invention occurred prior to the July 25, 2003

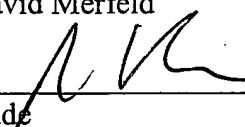
United States filing date of the application leading to U.S. Patent No. 6,835,786 B2 to Ishii et al.

7. The undersigned declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Date: \_\_\_\_\_ Kenneth Paul Zarnoch

Date: \_\_\_\_\_ John Robert Campbell

Date: \_\_\_\_\_ Glen David Merfeld

Date: 10/6/05 \_\_\_\_\_  
John Rude 

Date: \_\_\_\_\_ Prameela Susarla

Date: \_\_\_\_\_ Michael Alan Vallance

Date: \_\_\_\_\_ Gary William Yeager

O I P E  
PATENT  
OCT 14 2005

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EXHIBIT 1

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Date	Sample TRADEMARK OR DESCRIPTION	Spiral flow length, inches	Mold temp, oC	Top platen temp, oC	Plunger pressure x 0.05, psi	Amount of charge, g	
30-May	F 4	Base	15.5	129	140	43.5	15
31-May	F 4	Base	13	160		50	15
31-May	F 5	F4 with 0.30 IV PPO	7	160	168	50	15
31-May	F 5	F4 with 0.30 IV PPO	8.5	125	130	40	15
31-May	F 5	F4 with 0.30 IV PPO	8.5		145	50	15
7-Jun	F 8	Diff wax, less than F4	5	150		40	15
7-Jun	F 9	F8 with 1/2 initiator	4	150.5	150	40	15
7-Jun	F 10	F8 with no initiator	6.75	150.7	151.1	40	15
7-Jun	F 11	F8 with 1/2 PPO	11.3	149.8	149.9	40	15
7-Jun	F 12	F8 with added inhibitor	12	150.2		40	15
7-Jun	F 8		2	149.4	151.3	28	15
7-Jun	F 12	F8 with added inhibitor	11.3	150	156	50	15
7-Jun	F 12	F8 with added inhibitor	11	131.3	134.5	40	15

Sample	Temperatures, °C (Mold well)	Spiral Flow Length, inches	
Formulation 4	129-150	14-16	Base
Formulation 5	125-160	8.5	0.30 IV PPO
Formulation 8	150	5	Base + different wax, lower amount
Formulation 9	150.5	4	8 - with ½ peroxide
Formulation 10	150.7	6.75	8 - with no peroxide
Formulation 11	149.8	11.3	8 - with ½ PPO
Formulation 12	150.2	12	8 - with inhibitor

## EXHIBIT 1

page 2 of 3

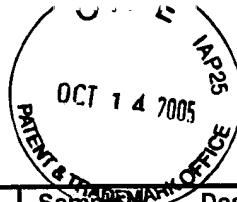
Plunger time, s	Clamp time, s
35	200
45	200
45	200
45	200
45	200
30	200
30	200
30	200
30	200
30	200
30	200
30	200

Formulation	4	5	8	9
PPO-MA(0.12)	8.7		8.99	8.99
PPO-MA(0.30)		6.1		
Diallylphthalate	8.6	11.2	8.89	9.07
TMPTMA	3.1	3.1	3.2	3.28
Dicumyl Peroxide	0.5	0.5	0.52	0.26
t-butyl catechol				
Lecithin	1.1	1.1		
Esterified Montanic Acid			0.2	0.2
Montanic acid			0.2	0.2
Dibromostyrene	1	1	1	1
Antimony pentoxide	1.3	1.3	1.3	1.3
Carbon black	0.2	0.2	0.2	0.2
Silane coupling agent	0.5	0.5	0.5	0.5
Fused silica	75	75	75	75
<b>Total</b>	<b>100</b>	<b>100</b>	<b>100</b>	<b>100</b>

# EXHIBIT 1

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10	11	12
8.99	4.5	8.94
9.27	12.19	8.89
3.34	4.39	3.2
0	0.52	0.52
		0.05
0.2	0.2	0.2
0.2	0.2	0.2
1	1	1
1.3	1.3	1.3
0.2	0.2	0.2
0.5	0.5	0.5
75	75	75
100	100	100



# EXHIBIT 2

page 1 of 2

Date	Sample	Description	Spiral flow length, inches	Mold temp, °C	Top platen temp, °C	Plunger pressure x 0.05, psi	Amount of charge, g	Plunger time, s	Clamp time, s
	F 4	Base	15.5	129	140	43.5	15	35	200
	F 4	Base	13	160		50	15	45	200
	F 5	F4 with 0.30 IV PPO	7	160	168	50	15	45	200
	F 5	F4 with 0.30 IV PPO	8.5	125	130	40	15	45	200
	F 5	F4 with 0.30 IV PPO	8.5		145	50	15	45	200
	F 8	Diff wax, less than F4	5	150		40	15	30	200
	F 9	F8 with 1/2 Initiator	4	150.5	150	40	15	30	200
	F 10	F8 with no Initiator	6.75	150.7	151.1	40	15	30	200
	F 11	F8 with 1/2 PPO	11.3	149.8	149.9	40	15	30	200
	F 12	F8 with added Inhibitor	12	150.2		40	15	30	200
	F 8		2	149.4	151.3	28	15	30	200
	F 12	F8 with added inhibitor	11.3	150	156	50	15	30	200
	F 12	F8 with added inhibitor	11	131.3	134.5	40	15	30	200

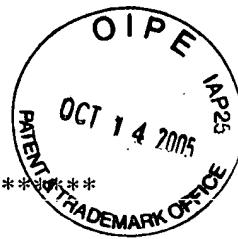
Sample	Temperatures, °C (Mold well)	Spiral Flow Length, inches	
Formulation 4	129-150	14-16	Base
Formulation 5	125-160	8.5	0.30 IV PPO
Formulation 8	150	5	Base + different wax, lower amount
Formulation 9	150.5	4	8 - with ½ peroxide
Formulation 10	150.7	6.75	8 - with no peroxide
Formulation 11	149.8	11.3	8 - with ½ PPO
Formulation 12	150.2	12	8 - with inhibitor

## EXHIBIT 2

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Formulation	4	5	8	9	10	11	12
PPO-MA(0.12)	8.7		8.99	8.99	8.99	4.5	8.94
PPO-MA(0.30)		6.1					
Diallylphthalate	8.6	11.2	8.89	9.07	9.27	12.19	8.89
TMPTMA	3.1	3.1	3.2	3.28	3.34	4.39	3.2
Dicumyl Peroxide	0.5	0.5	0.52	0.26	0	0.52	0.52
t-butyl catechol							0.05
Lecithin	1.1	1.1					
Esterified Montanic Acid			0.2	0.2	0.2	0.2	0.2
Montanic acid			0.2	0.2	0.2	0.2	0.2
Dibromostyrene	1	1	1	1	1	1	1
Antimony pentoxide	1.3	1.3	1.3	1.3	1.3	1.3	1.3
Carbon black	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Silane coupling agent	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Fused silica	75	75	75	75	75	75	75
<b>Total</b>	100	100	100	100	100	100	100

# EXHIBIT 3



\*\*\*\*\*  
GE Confidential & Proprietary Information.  
This invention is being prepared for submission  
to the GE Patent And Legal Operation. Attorney  
work product may be contained herein.

\*\*\*\*\*

## **GE Patent Disclosure Letter System**

### **DOCKET NUMBER**

31383

### **DOCKET DATE**

### **TITLE OF INVENTION**

Molding Material with High Mold Flow

### **GE TECHNOLOGY AREA(S)**

- GE Plastics (PLAS)
- GE Specialty Materials (GESM)

### **PROJECT NAME**

Engineering Composites

### **PROJECT NUMBER**

2140121001

### **PROJECT LEADER**

Vallance, Michael, A

### **BUSINESS OR ORG. CONTACT INFORMATION**

**NAME**

**PHONE NUMBER**

Was this invention first conceived or reduced to practice in  
the performance of work under a contract between GE and

# EXHIBIT 3

**another non-government third party? NO**

**Date Invention Conceived :**

**Circumstances Invention Conceived i.e., described in patent notebook (include page #), technical report, letter, discussed in meeting minutes, etc.**

We designed and executed an experiment to understand which factors in our molding compound compositions control flow length in the EMMI spiral flow test (SEMI G11-88 Recommended Practice for Ram Follower Gel Time and Spiral Flow of Thermal Setting Molding Compounds).

**Was this invention first conceived or reduced to practice in the performance of work under a US Government contract?**

NO

## **ABSTRACT OF THE INVENTION**

**Please write a brief explanation of the invention (Limit to 350 words)**

Molding materials which cure by free-radical polymerization can be tailored to provide maximum mold flow and minimum mold residence time.

## **BACKGROUND OF THE INVENTION**

**Please describe the problem or requirement addressed by your invention.**

During the encapsulation of electronic devices with molding compounds, low melt viscosity is required, often less than 10,000 cP for extended time, often for as long as 30 sec. Yet these same molding compounds are expected to cure sufficiently so as to attain hot hardness on the order of 90 or higher on the Shore D scale during short molding times, often as little as 90 sec.

**How has this problem or requirement been addressed before?**

Most molding compounds in use today comprise epoxy resin, polyphenol hardener, nucleophilic cure accelerator and high filler loading for control of thermal expansion. The organic compounds are chosen to provide low viscosity and slow cure. While sufficient polymerization occurs during the mold cycle to provide for mechanical integrity and sufficient hot hardness for removal of the molded components, extensive post curing, typically 5-6 hours at 175 C, is required to complete the curing process.

**Is this disclosure letter related to any GE disclosure letters, patent applications or issued patents?**

YES

# EXHIBIT 3

Vallance et al. re: PPO and colloidal silica Vallance et al. re:  
PPO and phosphorus flame retardant

**Have you completed a prior art search? NO**

**Please list any relevant literature or patents of which you are aware.**

## **DETAILED DESCRIPTION OF THE INVENTION**

### **How does your invention work?**

A curing chemistry is selected that results in addition polymerization kinetics. Such chemistry polymerizes rapidly and exothermically going to completion in a few seconds, provided there are no steric or diffusion limitations. While such choice of chemistry satisfies the need for rapid cure, spiral flow lengths are too short -- molding performance is unsatisfactory. We have discovered initiator strategies that overcome this shortcoming, providing rapid cure and long spiral flows.

### **Describe the important features of your invention and explain how to use the invention to solve the problems described above.**

In one aspect, the invention is a molding material. The material comprises: thermoplastic, filler, reactive diluent, and initiator. Other components may be present. The reactive diluent comprises a monomer with alkenyl functionality. The initiator comprises a free radical source and a free radical inhibitor. In a second aspect, the invention is a design paradigm. The paradigm comprises: Selection of a free radical source with a decomposition temperature very near to the molding temperature. Incorporation of an excess quantity of said free radical source. Incorporation of a free radical inhibitor in such quantity so that the spiral flow length is limited by the yield stress of the molding compound, which acts essentially as a Bingham fluid, rather than by the increase in viscosity associated with polymerization and/or crosslinking. Balancing the chemistry, so that the mold flow is essentially complete, before chain addition polymerization initiates, yet providing for polymerization at a rapid rate after onset of polymerization, similar to the rate of polymerization expected from an uninhibited system. Formation of a solid solution at room temperature, by the inclusion of a compatible high Tg thermoplastic, such that reaction is further impeded by diffusion control during storage and transport.

### **What advantages are provided by your invention?**

Chain-reaction cure kinetics are rapid in comparison to the polyetherification reaction associated with epoxy-phenolic cure, providing faster molding cycles, stronger as-molded

parts, and shorter post cures (post-cure-free performance in some circumstances). The polymers formed by addition polymerization of alkenyl functional monomers are generally less hydrophilic than the beta-hydroxy ethers formed during epoxy/phenolic cures. Moisture sensitivity is reduced. The inhibition time can be varied widely with subtle changes to the ratios in the initiator system, while impacting neither the polymerization rate nor the properties of the molded part. Therefore the chemistry is easily tunable to different molding geometries. The inclusion of a high-Tg thermoplastic results in improved toughness and, potentially, in lower moisture sensitivity, higher temperature capability, and better insulation properties, depending on the selection of polymer chemistry.

**Has your invention been reduced to practice? YES**

Date: 4/1/16

**Briefly describe any efforts to make a prototype of your invention or to test your invention. Additionally, summarize the results of any related experiments and testing and highlight any results of particular significance.**

We designed and executed an experiment to understand which factors in our molding compound compositions control flow length in the EMMI spiral flow test (SEMI G11-88 Recommended Practice for Ram Follower Gel Time and Spiral Flow of Thermal Setting Molding Compounds).

#### **BRIEF DESCRIPTION OF THE DRAWINGS**

**Please describe the significance of any pictures, drawings, graphs, diagrams, structures or figures and the type of picture along with the specific view or application to the invention.**

N.A.

#### **CLAIMED INVENTION**

**Please identify novel aspects that should be protected within this disclosure letter.**

(1) A molding material comprising (a) high Tg thermoplastic, (b) reactive diluent with alkenyl radicals, (c) free radical initiator, (d) free radical inhibitor, and (e) filler, where (i) decomposition temperature of the free radical initiator is near the molding temperature; (ii) the free radical initiator concentration is in excess of the amount required for polymerization of the reactive diluent; (iii) the free radical inhibitor concentration is based on attaining the desired spiral flow length at the selected molding temperature, typically 80-100 cm; and (iv) a solid or highly viscous solution is formed between the reactive diluent and some fraction of the thermoplastic. (2) The molding material of (1) wherein the

# EXHIBIT 3

thermoplastic is PPE with number average molecular weight from the range 2000 to 20,000. (3) The molding material of (2) wherein the phenolic hydroxyl radicals on the PPE are capped so as to prevent interference with the curing reaction and/or to enhance compatibility of the PPE with the remainder of the composition. (4) The composition of (1) wherein the reactive diluent cures through vinyl, allyl, acrylate or alkylacrylate radicals, but preferably acrylate or methacrylate. (5) The composition of (1) wherein the free radical source is a peroxide. (6) The composition of (5) wherein the peroxide is dicumyl peroxide or t-butyl peroxybenzoate. (7) The composition of (1) wherein the free radical inhibitor is a phenol or a hydroquinone. (8) The composition of (7) wherein the inhibitor is hydroquinone, hydroquinone mono-methyl ether, t-butyl hydroquinone, or t-butyl catechol. (9) The composition of (1) wherein the filler is silica. (10) The composition of (9) wherein the filler is essentially spherical. (11) The composition of (10) wherein the spherical filler is fused silica and/or colloidal silica. The filler may optionally be treated for improved compatibility with the remainder of the molding material and/or for reduced moisture sensitivity. (12) The composition of (1) wherein the filler is graded so as to essentially consist of particles smaller than a selected size, typically from the range 5 - 100 microns. (13) A process whereby the molding compound of (1) is molded into an article with the mold temperature selected from the range 140 to 170 C.

**ATTACHED FILES**

- (021016,Vallance) Inhibitor Study.ppt
- (021127,Susarla) Spiral flow study for Disclosure.xls

<b>DUTY OF DISCLOSURE</b>	
a.	Have steps been taken to put into use, either outside GE or in our own operations?
b.	Has the invention or a product embodying or using it been sold or offered for sale?
c.	If the invention pertains to a process, have any steps been taken to employ the process commercially (e.g., for product production)?
d.	Has the invention been described in an electronic or printed publication?
e.	Has the invention been described to persons who are not employees of GE?
f.	Are there results available of a prior art search

## EXHIBIT 3

	pertaining to this invention?	
g.	Has anyone else associated with the project within GE (marketing, sales, sourcing, etc.) disclosed the invention or offered the invention for sale?	No
h.	If you answered Questions a-g as "NO", is any use, sale, publication, or disclosure of the invention now contemplated?	No

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Kenneth Zarnoch	7 Horstman Drive Scotia, NY zip - 12302	Polymer & Specialty Chemical Technologies (2100)
Prameela Susarla	407 Lookout Lane Clifton Park, NY zip - 12065	Polymer & Specialty Chemical Technologies (2100)

\*Lead co-inventor

Primary / Financing Business (or Advanced Technology Program) : GE Plastics

Primary / Financing Component : Engineering Composites

Associated Lab/Program : Polymer & Specialty Chemical Technologies (2100)/Polymer Systems & Composites

Assigned Attorney : S. Brown